

## Comment on ‘Constant stress and pressure rheology of colloidal suspensions’.

In a recent Letter, Wang and Brady (WB) [1] analyse the rheology of Brownian hard spheres using constant stress and pressure Brownian dynamics simulations. The main observable is the shear viscosity,  $\eta(\dot{\gamma}, \Pi)$ , expressed as a function of the shear rate  $\dot{\gamma}$  and adimensional pressure  $\bar{\Pi} = \Pi a^3/(k_B T)$ , where  $\Pi$  is the pressure,  $k_B T$  the thermal energy, and  $a$  the average particle diameter. The central conclusion is the discovery of a “universal viscosity divergence” [1],

$$\eta \sim [\phi_m(\bar{\Pi}) - \phi]^{-\gamma}, \quad \gamma \simeq 2, \quad (1)$$

where  $\phi$  is the volume fraction and  $\phi_m(\bar{\Pi})$  a pressure-dependent critical density. WB argue that Eq. (1) is valid for all  $\bar{\Pi}$ , unifying the viscosity divergence of both thermal and athermal assemblies of hard spheres. Assuming that (1) describes the jamming transition of athermal hard spheres (at  $\bar{\Pi} \rightarrow \infty$ ), they conclude that the same physics must control the rheology at finite  $\bar{\Pi}$ . In this view, pressure only changes  $\phi_m(\bar{\Pi})$ , which interpolates between the jamming density  $\phi_J$  for  $\bar{\Pi} \rightarrow \infty$  and the glass transition density at low  $\bar{\Pi}$  [1], implying that Eq. (1) is valid also for the glass transition of Brownian hard spheres at equilibrium.

We show that these conclusions are not valid and provide the appropriate perspective to interpret WB’s results. We argue that the reported universality stems from exploring a single rheological regime where the hard sphere thermal glass is non-linearly sheared beyond yield, which directly explains the universal value  $\gamma \simeq 2$  in terms of the known hard sphere glass rheology, with no connection to the Newtonian regimes of either the colloidal glass transition or the granular jamming transition.

To assess the role of thermal fluctuations in hard sphere rheology, it is useful to consider not only the reduced pressure  $\bar{\Pi}$  but also the timescales associated with thermal fluctuations [2]. We consider two Péclet numbers:  $Pe_0 = \dot{\gamma} a^2/d_0$  and  $Pe = \dot{\gamma} a^2/d$ , where  $d(\phi)$  is the single particle diffusion coefficient at  $\dot{\gamma} = 0$ , and  $d_0$  its dilute limit. Only  $Pe$  is considered in [1], but because  $Pe_0 < Pe$ , three different regimes exist, which we use in Fig. 1 to organise the data of WB.

(i)  $Pe_0 < Pe \ll 1$ : The shear flow is slower than equilibrium relaxation ( $Pe \ll 1$ ). In this equilibrium regime, the viscosity is Newtonian,  $\eta = \eta_T(\bar{\Pi})$  [2]. It depends only on  $\bar{\Pi}$  and so cannot be varied in the constant- $\bar{\Pi}$  paths of WB. By construction, this approach cannot follow the rapid growth of the Newtonian viscosity of Brownian hard spheres on approaching the glass transition, which is indeed known [3] to differ from Eq. (1).

(ii)  $Pe_0 \ll 1 \ll Pe$ : This is the shear-thinning regime ( $1 \ll Pe$ ) of the thermal ( $Pe_0 \ll 1$ ) hard sphere glass. Most data described by (1) are in this regime (Fig. 1). WB’s universal collapse therefore describes the divergence of a *non-Newtonian* viscosity. These constant- $\bar{\Pi}$

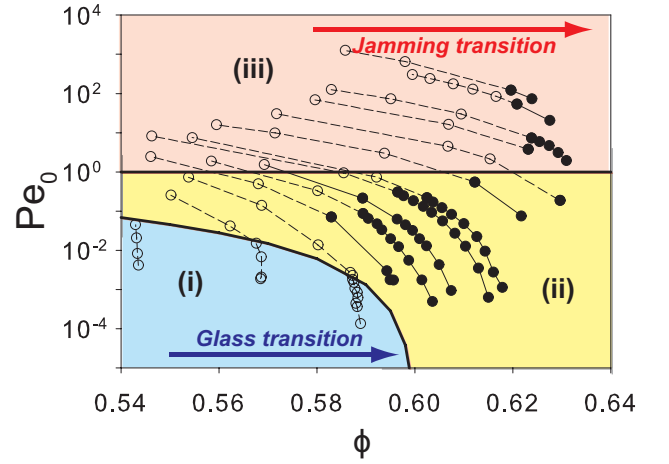


FIG. 1: Using adimensional shear rates, we organize the data of WB measured along constant- $\bar{\Pi}$  paths (dashed lines) into three regimes. Low viscosity data,  $\eta < 40$ , are shown as empty circles. (i) Newtonian thermal; (ii) shear-thinning thermal; (iii) Newtonian athermal. Filled symbols indicate data that are described by Eq. (1), having  $\eta > 40$ . As most of these data lie in (ii), Eq. (1) follows from the known rheology of the thermal hard sphere glass. The transition line between (ii) and (iii) is at  $Pe_0 = 1$ ; the one between (i) and (ii) ( $Pe = 1$ ) is determined by adjusting the viscosity model of [2] to WB’s data.

observations can be accounted for using known laws for the rheology of amorphous solids (including Brownian hard spheres). We assume Herschel-Bulkley rheology for both  $\sigma$  and  $\bar{\Pi}$ :  $\bar{\Pi}(\phi, \dot{\gamma}) = \bar{\Pi}_y(\phi) + b(\phi)\dot{\gamma}^n$ , where  $\bar{\Pi}_y(\phi)$ , the inverse function of  $\phi_m(\bar{\Pi})$ , is the pressure at yield. Expanding to linear order in density change at constant  $\bar{\Pi}$  gives  $\phi_m(\bar{\Pi}) - \phi(\dot{\gamma}, \bar{\Pi}) \sim \dot{\gamma}^n$ , thus  $\eta = \sigma/\dot{\gamma} \sim \sigma_y/\dot{\gamma} \sim [\phi_m(\bar{\Pi}) - \phi]^{-1/n}$ , showing that  $\gamma = 1/n$ . Numerical results are well described by  $n \simeq 0.5$  [2], in good agreement with the value  $\gamma \simeq 2$  reported by WB. Data in the inset of their Fig. 3a are also consistent with the known yield pressure divergence near jamming [4], from which we predict  $\phi_m(\bar{\Pi}) \sim \phi_J - c/\bar{\Pi}$  at large  $\bar{\Pi}$ .

(iii)  $1 \ll Pe_0 < Pe$ : Shear flow dominates even local thermal motion ( $1 \ll Pe_0$ ), so this regime is effectively athermal and the only one where jamming physics could be explored. The viscosity is Newtonian,  $\eta = \eta_0(\phi)$  and only depends on  $\phi$  or, in WB’s setup,  $\sigma/\Pi$ . As shown in Fig. 1 the data of WB are too sparse in this regime to assess the functional form of  $\eta_0(\phi)$ . Recent results show that Eq. (1) is again not valid asymptotically here [5].

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